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Author(s):

Russell D. Mosteller
David J. Loaiza
Rene G. Sanchez

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Russell D. Mosteller
Diagnostics Applications Groups (X-5)
Applied Physics Division
mosteller@lanl.gov

David J. Loaiza and Rene G. Sanchez Advanced Nuclear Technology Group (N-2) Nuclear Nonproliferation Division dloaiza@lanl.gov, rgsanchez@lanl.gov

Los Alamos National Laboratory

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ABSTRACT

Although neptunium is produced in significant quantities in nuclear power reactors, its critical mass is not well known. In addition, sizeable uncertainties exist for its cross sections. As an important step toward resolution of these issues, a critical experiment was conducted in 2002 at the Los Alamos Critical Experiments Facility. In the experiment, a 6-kg sphere of ²³⁷Np was surrounded by nested hemispherical shells of highly enriched uranium to produce a critical configuration.

Subsequently, a detailed analysis of that experiment has been performed using the MCNP5 Monte Carlo code in conjunction with a nuclear data library based on ENDF/B-VI. The results of that analysis are presented herein. It is found that a relatively small fraction (\sim 12%) of the fissions occur in the 237Np sphere and that criticality is underpredicted by \sim 0.01 Δ k. This is approximately three times the size of the underprediction for a bare sphere of highly enriched uranium. Consequently, it is concluded that better cross sections are needed for 237 Np.

ANALYSIS OF THE NP SPHERE EXPERIMENT WITH MCNP5™ AND ENDF/B-VI

Russell D. Mosteller
Diagnostics Applications (X-5)
Applied Physics Division
Los Alamos National Laboratory
mosteller@lanl.gov

David J. Loaiza and Rene G. Sanchez
Advanced Nuclear Technology (N-2)
Nuclear Nonproliferation Division
Los Alamos National Laboratory
dloaiza@lanl.gov,
rgsanchez@lanl.gov

Presented by Robert C. Little (X-5)

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BACKGROUND

Power reactors produce substantial quantities of ²³⁷Np and ²³⁹Np

²³⁹Np decays quickly ($t_{\frac{1}{2}}$ = 2.355 d), but ²³⁷Np has a very long half life ($t_{\frac{1}{2}}$ = 2.14 x 10⁶ yr)

²³⁷Np is produced by

- (n,2n) reactions on ²³⁸U
- (n,γ) reactions on ²³⁶U followed by β̄ decay of ²³⁷U

²³⁷Np has an effective threshold for fission at ~550 keV

Critical mass of ²³⁷Np is not well known, and neither are its cross sections

Previous estimates of the critical mass for ²³⁷Np have been based primarily on replacement measurements (tens of grams)



NP SPHERE EXPERIMENT

Experiment was performed in 2002 on the Planet vertical assembly machine at the Los Alamos Critical Experiments Facility

6.0704 kg sphere of nearly pure ²³⁷Np was surrounded by hemispherical shells of highly enriched uranium (HEU) to achieve criticality

Radius of Np sphere was 1.6335 inches (implied density is 20.289 g/cm³)

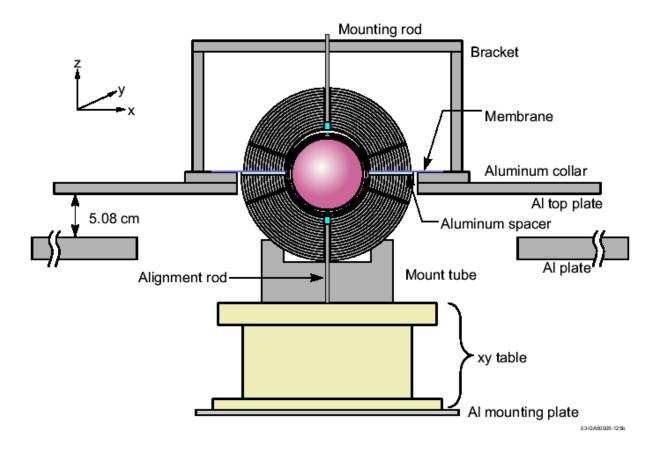
Sphere was enclosed in tungsten and two layers of nickel to reduce radiological dose from 310 keV γ rays from ²³³Pa

Total mass of HEU shells was 62.555 kg

Benchmark k_{eff} for detailed model of experiment is 1.0026 ± 0.0034



SCHEMATIC OF NP SPHERE EXPERIMENT





COMPOSITION OF NP SPHERE

| Element | <u>wt.%</u> |
|----------------------------------|-------------|
| Np (100% ²³⁷ Np) | 98.8 |
| U (78.7 wt.% ²³⁵ U) | 0.035 |
| Pu (88.2 wt.% ²³⁹ Pu) | 0.036 |
| Am (99.6 wt.% 243Am) | 0.183 |
| | |
| Total | 99.054 |

Sample assay did not dissolve completely, leaving 0.946 wt.% of the mass of the sphere unaccounted for



MCNP5 CALCULATIONS

MCNP5 calculations were performed to assess the impact of the "missing" mass of the sphere and to estimate k_{eff} for the experiment

Calculations employed a combination of the ACTI and ENDF66 continuousenergy nuclear data libraries, corresponding to ENDF/B-VI.8

Each calculation employed 650 generations of 10,000 neutrons each

First 50 generations were excluded from the statistics

Results therefore are based on 6,000,000 active neutron histories



REACTIVITY IMPACT OF "MISSING" MASS

| Missing mass | | Fission Fraction | |
|-------------------|---------------------|---------------------------|------------------|
| represented as | Δk | ²³⁷ N p | ²³⁵ U |
| Void | _ | 0.1264 | 0.8570 |
| Carbon | -0.0005 ± 0.0004 | 0.1257 | 0.8577 |
| Iron | -0.0002 ± 0.0004 | 0.1257 | 0.8576 |
| Europium | -0.0011 ± 0.0004 | 0.1258 | 0.8576 |
| Erbium | -0.0004 ± 0.0004 | 0.1259 | 0.8574 |
| ²³⁷ Np | 0.0012 ± 0.0004 | 0.1278 | 0.8556 |

Reactivity impact of missing mass is $\sim \pm 0.0012 \Delta k$

Benchmark k_{eff} for detailed model of experiment with "missing" mass represented as void becomes 1.0026 \pm 0.0036



RESULTS FOR DETAILED MODEL OF EXPERIMENT

| Parame | eter | Value |
|--------------------------------|-------------------|-----------------|
| k _{eff} | | 0.9900 ± 0.0002 |
| Fission Fraction, by Energy | Fast | 0.9474 |
| | Intermediate | 0.0526 |
| | Thermal | 0.0 |
| Fission Fraction, by Material | ²³⁷ Np | 0.1247 |
| | ²³⁵ U | 0.8587 |

For comparison, k_{eff} for Godiva (bare HEU sphere) is 0.9965 ± 0.0002 (-0.0035 Δk relative to benchmark value)

 $^{237}Np,$ with 1 fission in 8, therefore produces an underprediction of $\sim 0.009~\Delta k$

Based on these results, T-16 estimates critical mass of ²³⁷Np as ~ 57 kg



CONCLUSIONS

Reactivity impact of "missing" mass in Np sphere is small and can be accounted for by a slight increase in the uncertainty associated with $k_{\rm eff}$

Relatively small fraction of fissions occurs in Np sphere (~ 1 in 8)

Critical mass of ²³⁷Np is estimated to be ~ 57 kg

Based on ENDF/B-VI, calculated k_{eff} for this experiment is $\sim 0.0125~\Delta k$ low

²³⁷Np cross sections need to be improved

